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Final Report

1 Project Summary

Our research in the area of complex semiconductor structures has yielded substantial results that include better understanding of physical phenomena in quantized structures as well as a range of proposals for novel devices utilizing these effects. Among the effects studied are interface island formation, photoluminescence saturation due to traps, localization and delocalization and associated with it superluminescence, cascading of nonlinear effects in vertical structures, optical rectification, and coherent current control.

We propose to continue our research in the following directions: fabrication and testing of all-optical frequency shifters (and other cascading devices) for optical communications, further development of intersubband lasers, theoretical study of a fundamentally new type of low threshold "image" lasers, investigation of all-semiconductor magnetic materials, and new types of electronic sensors.

The results of our research shall contribute to both better understanding of physics of confined structures and to their practical application.

2 Introduction

Current research effort had been initiated by the AFOSR in May 1989 and since then it had been renewed twice. The main effort of the proposal has always been to conduct cutting edge theoretical and experimental research in the rapidly developing area of semiconductor nanostructures with an eye on potential applications in the areas of communications and information processing. In the mid-1980's semiconductor quantum wells (QW's) and superlattices (SL's) have been shown to possess electronic and optical properties dramatically different from those of bulk semiconductors. Immediately a number of practical proposals for novel electronic and electro-optical devices have been put forward. These devices included QW lasers, infrared detectors, high mobility transistors, tunneling devices and electro-optical modulators.

Our group's research has been focused on using the methods of "band-gap engineering" to improve various electronic and optical properties of materials. For instance, we have pioneered use of asymmetric QW's for second harmonic generation and other second-order nonlinear processes, we introduced the concept of the four-level intersubband laser, which contributed to the development of Quantum-Cascade Lasers. Idea of asymmetric SEED device was also suggested by our group. We also studied nonlinear optical processes in p-doped and type-II heterostructures, as well as the use of heterostructures for generation of THz microwave pulses. The emphasis of our work has always been placed on evaluating the feasibility of practical military and commercial applications.

In this proposal we first give a brief review of the major achievements of the third stage of our effort (1994-1996) and then outline the directions of future research.

In Section 3 we discuss the results of the research conducted in the course of two-and-a-half years elapsed since the last renewal of the project. From an experimental point of view, we have achieved the capability of routinely performing photoluminescence, photo-conductivity and photoluminescence excitation measurement at the JHU facilities. Femtosecond studies can be now conducted in collaboration with Dr. Y. Ding group at Bowling Green University, where a number of exciting experiments on the dynamics of optical processes involving interface islands have been carried out. We have also performed some experimental work in collaboration with NASA Goddard Space Center devoted to generation of $100\mu\text{m}$ radiation in QW's.

Among the most important theoretical results we describe advances in intersubband lasers and Raman oscillators, especially a new "inverted effective mass" scheme. We have also developed a theory of optical generation of THz radiation in bulk semi-

conductors and QW's and explained the experimental results of other groups. A major achievement has been the development of a rigorous theory for a group of phenomena commonly known as "lasing without inversion". For the first time we have developed expressions for threshold and slope efficiency and have come to conclusion that at least for our case of interest - quantum-confined semiconductor structures "lasing without inversion" does not offer any advantage over more conventional schemes. We also proposed a number of novel optoelectronic devices based on localization/delocalization transitions in superlattices and a fundamentally novel concept of low threshold lasing using image charges in quantum dots. We have fully developed a concept of using cascaded nonlinearities in vertical cavity structures to perform a number of standard nonlinear operations.

In section 4 we delineate general directions of the research for the next three years: it involves development of novel frequency shifters for WDM (4.1) and parametric oscillators (4.2), as well as further development in the area of intersubband lasers (4.3) directed toward reducing the threshold and increasing tunability in both longitudinal surface-emitting configurations. We plan to consider a fundamentally new type of low-threshold lasers based on image charges (4.4). All of the above directions are essentially a continuation of current efforts. There are also few fresh directions such as a proposal for all-semiconductor magnetic quantum dot arrays for memory and switching applications (4.5), research in the area of wide-gap semiconductors (4.6), and novel electronic devices and sensors (4.7).

Section 5 describes the resources available to us through our own facilities as well through an extensive list of established co-operative efforts of which we plan to take full advantage.

Last two sections describe the vita of the PI and the budget.

3 Results achieved in the last two-and-a-half years (Feb. 1994 - Aug. 1996)

3.1 Major highlights

- Obtained interesting results on photoluminescence dynamics in multiple QW's. These results include observation and interpretation of the spatially-localized band-gap renormalization and band-filling effects in the growth-interrupted two and three asymmetric-coupled quantum-well structures, and offer the evidence of heavy hole scattering in asymmetric coupled quantum wells.
- Attained time-resolved spectroscopy capabilities.
- Generated $10\mu W$ of Terahertz radiation using frequency-mixing in asymmetric QW's.
- Explained the mechanism for generation of THz radiation in bulk semiconductors and QW's. These results revealed for the first time the reason behind the sign reversal of THz signal observed in experiments.
- Proposed novel structures for the intersubband lasers and Raman oscillators.
- Developed rigorous theory of "inversion-less lasing" and showed its relation to Raman oscillations and its practical implications.
- Performed theoretical studies of optical localization/delocalization effects in superlattices and proposed a number of novel devices.
- Fully developed the concept of vertical cascaded nonlinear devices and progressed towards their experimental realization. Two optimized multilayer structures have already been designed and grown for implementing frequency doublers and parametric oscillators and amplifiers.
- Published in excess of thirty refereed papers and fifty conference proceedings.

Detailed description of our progress follows:

3.2 Intersubband lasers

This work has been focused on four major areas - a comparative analysis of the intersubband lasers and Raman oscillators, study of the influence of ionized and neutral impurities on inter-well tunneling in quantum wells, investigation of the prospects for surface-emitting lasers based on effective mass reversal in the valence bands, and, finally on the feasibility of ultra-low threshold quantum-box lasers based on image charges.

3.2.1 Comparative Analysis of Optically-Pumped Intersubband Lasers and Intersubband Raman Oscillators.

A new tunable source of far infrared radiation based on the phenomenon of intersubband stimulated electronic Raman scattering in semiconductor quantum wells was proposed by us. The threshold, efficiency, and tunability range of the proposed Raman oscillator were evaluated and found to constitute an improvement over those of optically-pumped intersubband lasers.

Results have been reported at the CLEO-95 conference and published in the Journal of Applied Physics [19].

3.2.2 Influence of the ionized impurities on the tunneling rates in semiconductor quantum wells

We studied theoretically impurity assisted tunneling in multiple quantum wells structures. We have shown that by strategically placing the impurities, it is possible to change the tunneling rates by as much as two orders of magnitude. This offers an opportunity for "engineering" structures with "customized" tunneling rates suitable for achieving intersubband population inversion and lasing. Experimental effort is under way.

Results have been reported at the CLEO-95 conference and published in the IEEE Journal of Quantum Electronics [24]

3.2.3 Silicon-based surface-emitting intersubband lasers based on effective mass reversal in the valence band of semiconductor quantum wells.

We introduced a fundamentally new concept for mid-IR lasers. This concept combines advantages of conventional semiconductor lasers and intersubband lasers. The

concept is based on the change of sign in the effective mass due to valence subband mixing. This concept will be thoroughly investigated and hopefully implemented in the next few years.

3.2.4 Novel type of quantum box intersubband lasers based on Stokes shift by image charges.

We considered theoretically a novel lasing scheme in semiconductor quantum boxes. Ultra-low threshold pumping power can be obtained due to development of image charges in surrounding layers. The impact of this scheme on intersubband lasing have been stressed. It has been shown that the scheme can also operate as a "phonon-pumped laser".

Results have been reported at two conferences and published in Applied Physics Letters [25].

3.3 Generation of THz radiation in bulk semiconductors and quantum wells.

3.3.1 Generation of 2.5THz radiation by difference frequency mixing in GaAs/AlGaAs multiple QW waveguides.

For the first time, using frequency mixing of two detuned CO_2 lasers in thick asymmetric quantum well devices we obtained in excess of $10\mu W$ of 2.5 THz coherent radiation. This research is conducted in collaboration with NASA Goddard Space Center. We utilized large (in excess of 100\AA) dipole moments associated with intersubband transition to achieve $\chi^{(2)}$ and anomalous dispersion in the vicinity of TO phonon resonance in order to achieve phasematching. We are currently working on achieving similar results using two semiconductor lasers.

Results have been presented at CLEO-95

3.3.2 Optical Rectification and Terahertz Emission in the Semiconductors Excited Above The Bandgap.

A rigorous theory of optical rectification in zinc-blende semiconductors has been developed. This theory combines the bonding orbitals representation of the electrons in the semiconductor with the band structure representation. It has been shown that when the semiconductor is excited above the absorption edge there is a strong resonant enhancement of the optical rectification signal and connected with it emission of

the terahertz radiation. Both the magnitude and the temporal characteristics of this signal are closely related to the intraband relaxation processes in the valence band.

The results have been reported in Journal of Optical Society of America B [9]

3.3.3 Dispersion and Anisotropy of the Optical Rectification in the zinc-blende quantum wells.

It has been demonstrated theoretically that optical rectification and difference-frequency generation nonlinear coefficients, known to exist in the asymmetric quantum wells for certain directions of growth do not vanish even in the symmetric quantum wells. The dispersion and anisotropy of the optical rectification coefficient has been evaluated and its magnitude was found to be one order larger than for the bulk zinc blende materials. This dramatic change has been shown to be caused by the lifting of the valence band degeneracy. Practical applications for generating terahertz radiation have been considered.

The results have been reported in Journal of Optical Society of America B [27]

3.3.4 Generation of low-frequency current and terahertz radiation using $\chi^{(3)}$ in semiconductors.

We investigated theoretically difference frequency mixing in bulk zinc blende or diamond structure semiconductors and shown that a lower-frequency (DC - 10THz) directional photocurrent and voltage can be generated as a result of the third-order nonlinear interaction. Practical applications for phase-sensitive detector have been considered.

Results have been reported in the International Journal of Nonlinear Physics [9] and presented at the QELS conference.

3.3.5 Current-induced second harmonic generation in semiconductors

Direct current in semiconductor has been theoretically shown to be capable of doubling the frequency of incoming optical radiation. The second order susceptibility, proportional to the current is calculated to be in the $10^{-14} - 10^{-13} m/V$ range. Applications of the novel phenomenon to the probing and mapping of the current in semiconductor devices were considered.

The results have been reported in Applied Physics Letters [13]

3.4 Novel phenomena associated with localization and de-localization in the semiconductor superlattices.

3.4.1 Optical Wannier-Stark Effect

We have discovered a novel effect, consisting of optically-induced effective mass change due to the change in the degree of localization in the semiconductor superlattices. This effect can be manifested by change in conductivity caused by the below-the-gap optical radiation. Possible application as a non-absorbing differential light detector/switch is considered.

The results have been reported in Applied Physics Letters [5] and Optics Communications [11]

3.4.2 Controlled Super-luminescence in Semiconductor Superlattices

Radiative recombination of excitonic states in semiconductor superlattices in the presence of an electric field has been studied theoretically. It has been shown that when the electron-hole Coulomb interaction energy exceeds the miniband width, a coherent excitonic state is created whose oscillator strength surpasses the oscillator strength of a single quantum well by orders of magnitude. We also demonstrated that a small external field can split the coherent state into isolated well states and thus severely deplete the oscillator strength of the exciton. This opens the possibility of modulating and switching of super-radiance in semiconductor devices.

The results have been reported in Applied Physics Letters [12] and in [22]

3.5 Prospects for inversion-less lasers in semiconductor structures

A rigorous density matrix-based theory of lasing based on optically- or autoionization-induced coherence between energy levels has been developed. For the first time the balance equations have obtained, and the conditions for lasing threshold established as a function of pumping strength and relaxation rates. Connection between Lasing Without Inversion and Raman or parametric processes has been clearly established. The main conclusion is that true CW lasing without inversion can indeed be obtained but only in a system where pumping and relaxation rates are favorable for

attaining either conventional lasing with population inversion or for the stimulated Raman oscillations (Stokes and Anti-Stokes). Practical implications, especially for the intersubband lasers have been discussed.

Results have been published in Physical Review A [28,30] and on the IEEE Journal of Quantum Electronics [29].

3.6 All-Optical Switching and Frequency-Conversion Components Based on Cascaded Second-Order Nonlinearity

Many nonlinear optical phenomena depend on a nonzero value for the third-order nonlinear susceptibilities, $\chi^{(3)}$, which is very small in most conventional materials. Recently, it has been found that a cascaded second-harmonic generation (SHG) scheme could be used to effectively produce an effective third-order nonlinearity. The most effective SHG scheme uses surface-emitting SHG confined to a waveguide structure. Efficiency can be further improved by using the large second-order optical susceptibilities found in asymmetric quantum wells where the sign of $\chi^{(2)}$ can be modulated.

We proposed a family of all-optical switching devices and frequency conversion components based on cascaded second-order nonlinearity. All of these devices are based on the large $\chi^{(2)}$ of asymmetric QW's and characterized by low absorption, high efficiency, versatility, and the ability to be integrated with other opto-electronic components on the same chip. The technological steps involved in the fabrication of these devices are practically the same ones involved in fabrication of VCSEL's and DFB lasers.

The proposed devices are:

- Optical limiter
- Self-phase modulator
- Optical frequency shifter
- Optical parametric oscillator
- Optical phase conjugator
- Nonlinear directional coupler

The results have been reported in a number of papers [2,10,32,35].

3.6.1 Surface-emitting second-harmonic generation

Following the quasi-phase matching scheme by spatially modulating second-order susceptibility, the surface-emitting second-harmonic generation (SHG) based on semiconductor multilayers or asymmetric coupled quantum-well domain structures has been investigated [Normandin et al Electr. Lett., 26, 2088,1990], but no definite results for maximum conversion efficiency have been reported.

We have systematically investigated the SHG under wide range of pump powers, and shown that when the pump power density is equal to saturation power density, the conversion efficiency reaches 72%. We have also predicted that in the surface-emitting geometry, when the vertical cavity is introduced, large field enhancement results, and, subsequently, the saturation power density is greatly reduced. Therefore, high conversion efficiency can be achieved at relatively low input power density.

Our calculations have shown that for on ZnSe/ZnS or GaAs/AlGaAs materials, the saturation power density is expected to be in the range of 0.1 - 5 / μm for the wavelength range of the fundamental beam: 1 - 2 μm . We also predicted that surface-emitting geometry offers a much wider usable bandwidth..

We have already designed and grown an optimized structure in collaboration with Army Research Labs. Currently, we are testing the performance of this structure as an efficient frequency doubler. Such an investigation is essential for all the devices discussed below.

Our results were published in [14-16,31-33].

3.7 Experimental work on dynamics of recombination in coupled quantum wells.

3.7.1 Spatially-localized band-gap renormalization and band-filling effects

We studied a number of multiple quantum wells structures:

- Three-QW structure with the growth interrupted at every interface.
- modulation-doped three-well structure.
- narrow asymmetric two-QW structure grown at NRL at temperature of 600C and with the growth interrupted for 60 seconds at every interface

- similar sample grown at UCLA with the growth interrupted for 10 minutes at each interface

Both CW and time-resolved experiments were performed. We observed a number of interesting phenomena including pronounced broadening of the photoluminescence linewidth caused by the increase in laser intensity. This broadening, which had not been observed before, was caused by spatially-localized band-gap renormalization. The explanation for it is as follows: because of the growth interruption at each interface, one peak in photoluminescence spectrum breaks into two. They correspond to recombination of localized excitons at interface islands as well as recombination of free excitons.

In the modulation-doped three-well structure, the photoluminescence was shown to be dominated by impurity emission at low intensity. However, as the intensity increases, the emission peak due to the recombination of the spatially-localized excitons dominates. As the intensity increases further, free-exciton recombination dominates. We believe we have almost completely filled the impurity sites and localized exciton states sequentially (band-filling effect).

Time-resolved experiments have allowed us to measure the recombination times in modulated and undoped structures. The observed difference has been attributed to free exciton screening.

Results have been reported in JOSA B and other publications [2,8,20]

3.7.2 Scattering of heavy holes

In an asymmetric-coupled quantum-well structure, we have observed evidence of heavy-hole tunneling based on CW PL via acoustic phonons. When the sample temperature is below 80 K, the heavy-hole associated with the wide well is scattered into the narrow well at high energy subbands. On the other hand, when the temperature is above 100 K, the heavy hole in the narrow well is scattered into the wide well at the top of the energy band. In the second asymmetric-coupled quantum-well structure, we have observed evidence of the optically-induced redistribution of the electron-hole pairs by studying CW photoluminescence spectra. As the laser intensity increases, the ratio of the first two heavy-hole emission intensities increases. Our preliminary analysis shows that this is the result of the scattering of heavy holes between two coupled quantum wells, strongly affected by the spatial charge separation between optically-generated electrons and heavy holes.

Our results were presented at ILS Conference .

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14. Y. J. Ding, J. B. Khurgin, and S. J. Lee, "Transversely-Pumped Counter-Propagating Optical Parametric Oscillators and Amplifiers: Conversion Efficiencies and Tuning Ranges" *IEEE J. Quan. Electron.* **QE-31**, 1648 (1995).
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33. Y. J. Ding and J. B. Khurgin, "Quasi-phase-matched mirror-less backward optical parametric oscillation" submitted to *Phys. Rev. Lett.*,
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